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M. DUTT, J. A. ELLIOTT, B. C. HANCOCK and A. C. BENTHAM

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Granular templating: Effects of boundary structure on particle packings under simultaneous shear and compression

M. Dutt[1], J. A. Elliott[1], B. C. Hancock[2] and A. C. Bentham[3]

[1] *Department of Materials Science and Metallurgy, University of Cambridge - Pembroke Street, Cambridge CB2 3QZ, UK*
[2] *Pfizer Global Research and Development - Groton, CT 06340, USA*
[3] *Pfizer Global Research and Development - Sandwich, Kent, CT13 9NJ, UK*

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Abstract – We present our findings on the effect of various confining substrates, both crystalline and amorphous, on spherical particles, packed under gravity followed by the simultaneous application of shear and compressive strains. We treat the voids and particles of the substrates interchangeably by identifying the primitive unit cell for each, and use radial and angular distribution functions to determine the packing structures. We show that a substrate templated with a 2D square lattice, for which void and particle packing structures are identical, is most suitable for inducing crystallisation mimicking the substrate structure.

Boundary conditions in most laboratory and numerical experiments studying the behaviour of granular materials are set up with immense care so as to not unduly influence the phenomenon under scrutiny. However, if one wants to explore the effect of boundaries on the generation of ordered packing structures, independent of the inter-particle interaction forces, then templated substrates must be utilised. Studies on colloidal systems [1–3] have explored the crystallisation of colloidal particles using templated surfaces via various intermolecular and surface interactions due to their potential use in sensors and photonic switches. Applications in tissue engineering often require scaffolds formed from networks of regular arrays of pores or struts so as to utilise the cavities for directed tissue growth [4]. In addition, systematic studies of fluid flow through porous media requires regular arrays of cavities in the porous scaffolds in order to obtain comparisons with theoretical predictions [5,6]. Studies on hard disks and spheres [7,8] have addressed similar queries for non-cohesive hard macroscopic particles.

We probe the effects of 2D crystalline and random substrate morphologies on the 3D packings of dry non-cohesive granular particles confined between substrates. These particles interact via dissipative contact forces, with mechanical excitation and external forces such as gravity being the only means to achieve spatial rearrangement. We use three different confining substrates generated from monodisperse spheres either arranged in a 2D square (*i.e.*, simple cubic (s.c.) (100)) or triangular (*i.e.*, hexagonal close-packed (h.c.p.) (001)) lattice, shown as insets in fig. 1, or fused randomly. We initiate the particle packing process by allowing the particles to settle under gravity onto each substrate. We then impose external fields which induce the system to crystallise using highly conducive conditions, such as uniaxial compression and shear, applied parallel and perpendicular, respectively, to the direction of gravity. The compressive strain (rate $\dot{\gamma_c}$) pushes the particles towards the bounding substrates, filling up the neighbouring voids. The shear strain (rate $\dot{\gamma_s}$), applied simultaneously with the compression, allows the particles to leave their constrained states so as to explore more globally stable configurations, and thereby influences the void filling process by enabling the particles to explore the neighbouring interstices and translating the applied shear to the particles in the layers below. We observe that the s.c. (100) substrate is responsible for seeding the growth of a 3D ordered packing while the particles settle under gravity. We propose that a combination of the

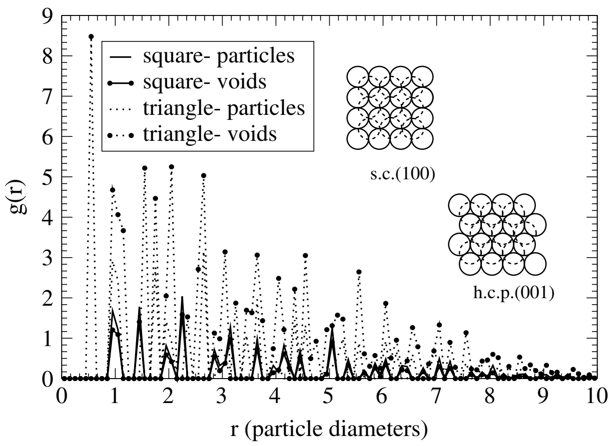

Fig. 1: Calculations of $g(r)$ for void and particle pairs. The insets are, respectively, the s.c. (100) and h.c.p. (001) substrates with the markings indicating the void PUC, in solid lines, and the optimal occupancy of the first layer, in dotted lines.

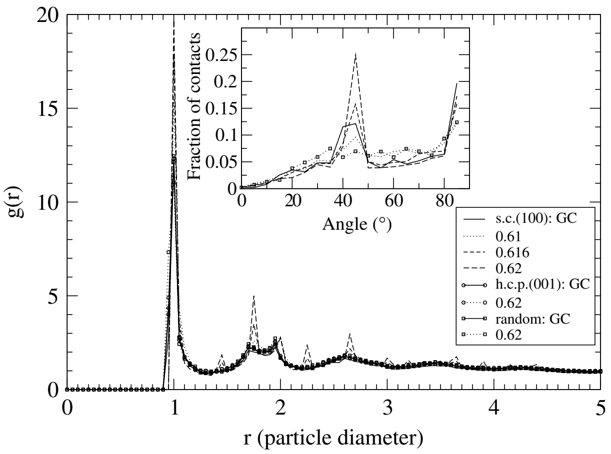

Fig. 2: Calculations of $g(r)$ and contact angle distribution (inset) for $\dot{\gamma_s} = 0.00399$ and $\dot{\gamma_c} = 0.0399$; GC is gravity compaction.

initial random spatial configuration of the particles, the interparticle interactions and the simultaneous occupancy of all voids on the s.c. (100) substrate, as opposed to the alternate simultaneous occupancy of voids of the h.c.p. (001) substrate, is responsible for this result.

The particles, modelled using the mechanical properties of microcrystalline cellulose (Young's modulus of 9.08 GPa [9] and the Poisson's ratio of 0.3 [10]), were employed in the 3D numerical experiments with 2D periodic boundary conditions, which were carried out using DEM simulations [11]. A system of 1800 spheres was constrained along the vertical direction by two bounding substrates; the substrate particles were identical to those in the bulk. Hertzian contact laws along with Coulomb's yield criteria were used to model the inter-particle contact interactions; the coefficients of kinetic and static friction were set to 0.1. Homogenisation of the initial particle positions occurs via elastic hard-sphere interactions for a given number of cumulative collisions. The numerical experiment is carried out in two phases, both occurring over fixed intervals of time. First, the particles are allowed to settle under gravity, then, while the settling process is occurring, the upper substrate begins to apply compressive and shear strains at constant rates $\dot{\gamma_c}$ and $\dot{\gamma_s}$, respectively. The simulation cell dimensions are determined by the height of the cell (parallel to the direction of gravity and the applied compression), and the dimensions of the bounding substrate, which is measured from the centres of mass of the substrate particles located at its extremities. The simulation cell dimensions remain unchanged throughout the simulation; however during the strain application phase (SAP), the height of the upper bounding substrate reduces at a rate proportional to the compressive strain rate while the magnitude of the difference in the system particle positions, at the extremities, along the x- and y-directions increases negligibly so as to allow the particles to rerrange themselves. The latter is allowed on account of two-dimensional periodic boundary conditions along the x- and y-directions.The SAP will last as long as each particle does not deform by more than 1% of its diameter. The simulation stops when the deformation criterion fails. To obtain a comparison between the effects of gravity and the compressive strain, we use the particle diameter a, mass m and gravity g to develop a system of units. The compressive and shear strain rates, measured in units of $\sqrt{2ag}$, lie in the range 0.00399–0.0439 and 0.00399–0.00798, respectively. The time unit is taken to be that for a particle to fall through a distance equivalent to its diameter, *i.e.*, $\sqrt{2a/g}$ and therefore, the integrating time step is approximately 1.5×10^{-5}. The integrating time step has been chosen such that the particles undergo quasi-static compression for all values of the compressive strain. Using the maximum value of $\dot{\gamma_c}$ (= 0.0439), the integrating time step is set such that at each iterative step, the compressive substrate moves by a distance of $10^{-4}a$. The stress unit is given by mg/a^2.

We explore whether the degree of similarity between the structure of the void and particle lattices is an indication of the suitability of the substrate to induce crystallisation such that the packing structure emulates that of the substrate. Figure 2 shows calculations of the pair correlation function $g(r)$ and the fraction of contacts with a given contact angle ϕ (measured between the inter-particle contact vector and the gravity axis) for bulk particles, before and after the SAP, for different instances (*i.e.*, packing fractions) of the numerical experiments carried out with the three substrates for strain rates $\dot{\gamma_s} = 0.00399$ and $\dot{\gamma_c} = 0.0399$. For a given bounding substrate, the packing characteristics of the particles before the SAP are identical. For the random and h.c.p. (001) substrates, the $g(r)$ curves indicate local close packing with the splitting of the second peak at inter-particle distances of $\sqrt{3}a$ and 2a. The distance $\sqrt{3}a$ is the longer diagonal of the packing of four particles located at the corners of a rhombus with side length a, whereas the distance $2a$ arises from the lineup of 3 particles. The fraction of contact angles remains invariant over the interval $25^\circ < \phi < 85^\circ$, with a preference for $\phi \sim 85^\circ$ due to the approximate layering of the particles. The s.c. (100) substrate also demonstrates the layering effect with some particles supported by four others located at the vertices of a square (such as those found in the lower substrate). On completion of the SAP, only the packings bound by the s.c. (100) substrate show the development of long-range order in the spatial configuration. This is on account of the preferred inter-particle distances of a, $\sqrt{2}a$, $\sqrt{3}a$, $2a$ and so on; the peak at $\sqrt{2}a$ arising due to the diagonal of a square which has particles at its vertices, in contact with one another. In addition, there is an emergence of highly preferred contact angles at 45° and 85°. These results strongly suggests successive interdigitating s.c. (100) layers developing through the particle packing.

We compare the substrate morphologies by treating the particles and voids interchangeably, *i.e.*, by considering the lattice generated by voids or particles. As a first approximation, we consider the 2D projection of each substrate. The traditional definition of a primitive unit cell (PUC) [12] can be extended to a *void* PUC defined such that it contains a single void (as marked in fig. 1). The total, occupied and unoccupied areas of the PUC for the s.c. (100) (h.c.p. (001)) lattice are $4a^2$ ($2\sqrt{3}a^2$), πa^2 (πa^2) and $(4-\pi)a^2$ ($(2\sqrt{3}-\pi)a^2$). Hence the particle and void area fraction for the s.c. (100) (h.c.p. (001)) lattice are 0.7853 (0.9069) and 0.2146 (0.0931). To obtain a more accurate estimate of the void and packing fractions of each substrate, we consider the 3D structure of each regular substrate. The total, occupied and unoccupied volumes (marked in fig. 1) of the s.c. (100) (h.c.p. (001)) substrate are $4a^3$ ($2\sqrt{3}a^3$), $(2/3)\pi a^3$ ($(2/3)\pi a^3$) and $(4-(2/3)\pi)a^3$ ($(2\sqrt{3}-(2/3)\pi)a^3$). Therefore, the packing and void fraction of the s.c. (100) (h.c.p. (001)) substrate are 0.5235 (0.6046) and 0.4767 (0.3954). Calculations of $g(r)$ for the void and particle pairs for both the lattices (fig. 1) demonstrate significant differences for the h.c.p. (001) substrate but none for the s.c. (100) substrate. A rough calculation of the first, second and third nearest inter-void (particle) neighbours for the h.c.p. (001) substrate are $a/\sqrt{3}$ (a), a ($\sqrt{3}a$) and $2a/\sqrt{3}$ (2 a), respectively. A similar set of calculations for the s.c. (100) substrate yields identical results for the inter-void (particle) neighbours. As a consequence, during the gravity packing phase, all the voids in the s.c. (100) substrate can be occupied by particles to form another layer which closely emulates the substrate lattice but shifted by half a lattice vector. In a h.c.p. (001) substrate, however, no two neighbouring voids can be simultaneously occupied by particles due to steric constraints. Similarly, the distribution of the angle between the vector joining the centres of mass of two particles and the x-axis for all the distinct combinations of the void-void and particle-particle pairs demonstrates a remarkable similarity only for the s.c. (100) substrate.

The impact of the substrate templated with the s.c. (100) lattice during the SAP can be tracked by determining the preferred interparticle distances and contact angles in different vertical sections of the system. For low values of $\dot{\gamma_c}$, the particles close to the lower substrate loosely adhere to the template of the s.c. (100) lattice, evolving to random close packing in the middle of the system, and finally to a random loose packing at the top of the system, as the upper substrate is in contact with a small fraction of the particles in the upper layer. For high values of $\dot{\gamma_c}$, similar results are obtained for the lower part of the system; however, contact of the upper substrate with an increasing number of particles leads to the development of successive interdigitating s.c. (100) layers which eventually grow from particle layers in proximity to each substrate to those in the middle of the system. In addition, the onset of the ordered packing occurs earlier with increasing values of $\dot{\gamma_c}$.

Due to the higher porosity of the s.c. (100) substrate, the particles, under compressive and shear strains, can fit into the voids on the substrate, or the neighbouring particle layers, thereby *seeding* successive interdigitating s.c. (100) layers. The lower void volume available in the h.c.p. (001) substrate coupled with the simultaneous non-occupancy constraint of the neighbouring voids and the initial random spatial configurations of the particles results in the inability of the particles to fit into the substrate voids, so as to begin propagating the substrate template and consequently, results in a frustrated random close packed configuration. In the presence of a cooperative occupancy of the interstices on the substrate layer, it is however possible to crystallise a system of particles compressed against a triangular particle bed. In addition, for small systems, the boundary conditions both in terms of finite-size effects and the shape of the simulation cell can stabilise certain types of ordered packing. However, for the systems that we have studied, the number of particles and the dimensions of the simulation cell are too large to be affected by such artefacts. The influence of the substrate effects on particle packings has been extended to other physical systems where free volume constraints along with intermolecular and surfaces forces dominate the dynamics [1–3].

We obtain a macroscopic quantification of the changes in the particle agglomerate, during the SAP, from the measurements of the shear stress (fig. 3, shown in non-dimensional units) and the packing fraction for the three substrates. The upper boundary does not come into contact with the settled particle agglomerate when the SAP commences as some of the particles are still settling under gravity. However, when contact is established, the combined action of the compressive and shear strains allows significant rearrangement in the particle packing, causing a jump in the densification and a linear depth profile of the shear stress for packing fraction $\rho \sim 0.59$–0.60. The resulting densification of the agglomerate causes the upper substrate to lose contact with

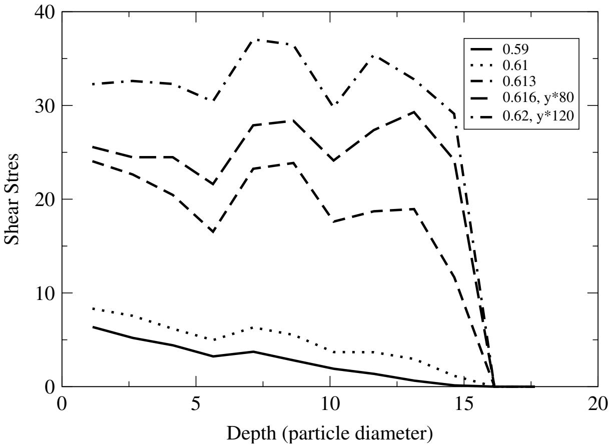

Fig. 3: Calculation of the shear stress, in non-dimensional units, as a function of depth for different instances (packing fractions, as indicated in the legend) of a numerical experiment carried out with the s.c. (100) substrate for $\dot{\gamma_s} = 0.00399$ and $\dot{\gamma_c} = 0.0399$.

the particles for an interval of time ($\propto 1/\dot{\gamma_c}$). As the upper substrate regains contact of the particles, the shear stress again develops a linear profile with the depth of the agglomerate for $\rho \sim 0.61$. For higher values of ρ, the shear stress increases in a manner so as to gradually lose its uniformly decreasing profile across the depth of the agglomerate, dropping off in the vicinity of the lower substrate. For sufficiently high densification of the agglomerate, we would expect the shear stress to develop a uniform depth profile as the upper substrate drags the agglomerate with it as if it were a solid mass.

We have described the generation of a granular crystal via gravitational deposition of the particles on a templated substrate (s.c. (100)), followed by the simultaneous application of compressive and shear strains. The similarity between the void and particle packing structures on the s.c. (100) substrate coupled with the nature of inter particle interactions and the application of vertical and lateral strain makes this substrate most conducive for seeding crystallisation in comparison to the h.c.p. (001) substrate. In spite of differences in packing structure generated by the three substrates, the time evolution of the stress profile across the sample is qualitatively similar. We find the results to be insensitive to the values of the shear strain rates indicating the dominance of the compression strain. We also observe that numerical experiments with smaller number of particles have shown the collective particle dynamics to evolve into inter-digitating s.c. (100) layers. The ordered pattern deviates from a body centred cubic (b.c.c.) packed structure due to contact between the spheres along edges and diagonals of the cube. For the b.c.c. lattice, only spheres along the body diagonal touch, not along the edges. In this letter, we have demonstrated that by using a numerical experiment, implemented by a fixed time dynamics integrated technique, where both lateral and orthogonal strains are applied to templated substrates for investigating the influence of a templated substrate on dense sphere packings, our results are in good agreement with those by Stillinger and Lubachevsky [7]. This, in turn, emphasizes the robustness of the phenomena of inducing ordered packings via the utilisation of suitably templated substrates. Our results also highlight a method to develop ordered packings of macroscopic particle ($\sim 200\,\mu$m) which can be used for applications which require periodic scaffolds or porous media.

* * *

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